AD-A066 284 FOREIGN TECHNOLOGY DIV WRIGHT-PATTERSON AFB OHIO F/G 11/2
MOSI2-GLASS COATINGS FOR SILICON CARBIDE ON A NITRIDE BINDER,(U)
MAR 78 M V SAZONOVA, Y A KARPICHENKO

UNCLASSIFIED FTD-ID(RS)T-0312-78 NL

1 OF 1
ADA
066284

END
DATE
FILMED
5-79
DDC

FTD-ID(RS)T-0312-78

1

FOREIGN TECHNOLOGY DIVISION

$MoSi_2$—GLASS COATINGS FOR SILICON CARBIDE ON A NITRIDE BINDER

by

M. V. Sazonova, Ye. A. Karpichenko

EDITED TRANSLATION

FTD-ID(RS)T-0312-78 15 March 1978

MICROFICHE NR: FTD-78-C-000438

$MoSi_2$—GLASS COATINGS FOR SILICON CARBIDE ON A NITRIDE BINDER

By: M. V. Sazonova, Ye. A. Karpichenko

English pages: 9

Source: Zashchitnyye Vysokotemperaturnyye Pokrytiya. Trudy Vsesoyuznogo Soveshchaniya po Zharostoykim Pokrytiyam, Khar'kov, 12-16 May 1970, Leningrad, 1972, pp. 166-173.

Country of origin: USSR
Translated by: Charles T. Ostertag, Jr.
Requester: AFML/LTM

PREPARED BY:

TRANSLATION DIVISION
FOREIGN TECHNOLOGY DIVISION
WP-AFB, OHIO.

U. S. BOARD ON GEOGRAPHIC NAMES TRANSLITERATION SYSTEM

Block	Italic	Transliteration	Block	Italic	Transliteration
А а	*А а*	A, a	Р р	*Р р*	R, r
Б б	*Б б*	B, b	С с	*С с*	S, s
В в	*В в*	V, v	Т т	*Т т*	T, t
Г г	*Г г*	G, g	У у	*У у*	U, u
Д д	*Д д*	D, d	Ф ф	*Ф ф*	F, f
Е е	*Е е*	Ye, ye; E, e*	Х х	*Х х*	Kh, kh
Ж ж	*Ж ж*	Zh, zh	Ц ц	*Ц ц*	Ts, ts
З з	*З з*	Z, z	Ч ч	*Ч ч*	Ch, ch
И и	*И и*	I, i	Ш ш	*Ш ш*	Sh, sh
Й й	*Й й*	Y, y	Щ щ	*Щ щ*	Shch, shch
К к	*К к*	K, k	Ъ ъ	*Ъ ъ*	"
Л л	*Л л*	L, l	Ы ы	*Ы ы*	Y, y
М м	*М м*	M, m	Ь ь	*Ь ь*	'
Н н	*Н н*	N, n	Э э	*Э э*	E, e
О о	*О о*	O, o	Ю ю	*Ю ю*	Yu, yu
П п	*П п*	P, p	Я я	*Я я*	Ya, ya

*ye initially, after vowels, and after ъ, ь; e elsewhere.
When written as ë in Russian, transliterate as yë or ë.

RUSSIAN AND ENGLISH TRIGONOMETRIC FUNCTIONS

Russian	English	Russian	English	Russian	English
sin	sin	sh	sinh	arc sh	$\sinh^{-1}$
cos	cos	ch	cosh	arc ch	$\cosh^{-1}$
tg	tan	th	tanh	arc th	$\tanh^{-1}$
ctg	cot	cth	coth	arc cth	$\coth^{-1}$
sec	sec	sch	sech	arc sch	sech^{-1}
cosec	csc	csch	csch	arc csch	csch^{-1}

Russian	English
rot	curl
lg	log

$MoSi_2$—GLASS COATINGS FOR SILICON CARBIDE ON A NITRIDE BINDER

Sazonova, M. V. and Karpichenko, Ye. A.

Silicon carbide and materials obtained on a base of it are oxidized in air at high temperatures with the formation on their surface of a silica film which is inclined to crystallization. Polymorphous transformations of crystalline silica, accompanied by major volumetric changes, promote the breakdown of silicon carbide [1, 2].

Methods are known for protecting heaters made of silicon carbide from oxidation with the help of coatings of $MoSi_2$ which are obtained in argon at 1500°. Coatings made from $MoSi_2$ increase their period of service by four times [3]. Coatings made from $MoSi_2$ and alkali-free borosilicate glass, which were obtained in the air, give reliable protection to graphite which has been siliconized from the surface at 1400-1500° [4].

We undertook an attempt to protect silicon carbide on a nitride binder from oxidation at 1500° in the air with the help of coatings consisting of $MoSi_2$ and refractory, highly siliceous boron-free glass.

CHARACTERISTICS OF THE INITIAL SAMPLES. Samples made out of SiC on a nitride binder were prepared by the All-Union Insitute of Refractory Materials. They contained (wt.%): SiC - 65, Si_3N_4 -

35, they had a porosity (apparent) of an order of 18-19% and a coefficient of thermal expansion (KTR), equal to $45 \cdot 10^{-7}$ 1/deg in the area of 20-800°. Heat treatment of the samples in the air was accompanied by an increase in their weight. At 1300° for 20 hours the increase of weight comprised 1.8%, at 1400° for 60 hours - 3.65%.

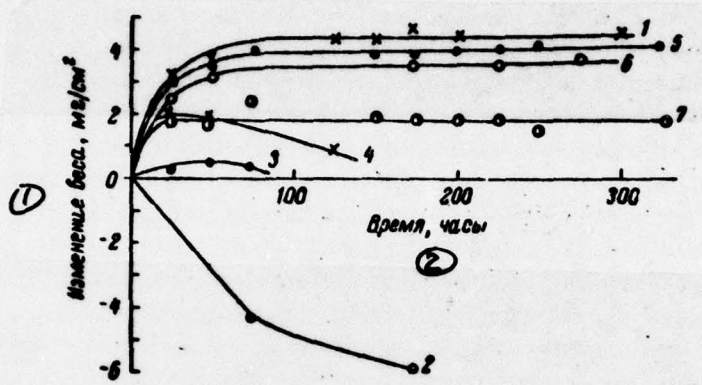

Figure 1. Oxidation of silicon carbide on a nitride binder at 1500° in the air.
1, 2 - without a coating; 3-5 - with coatings Nos 3, 5, 6 respectively; 6, 7 - with double-layer coatings Nos 7, 8.
Key: (1) Change in weight, mg/cm^2; (2) Time, hours.

With in increase in the temperature of testing a scattering of data on the heat tolerance of this material was observed. At a temperature of 1500° samples from the same batch reacted differently (Figure 1, 1,2). The increase in weight of some of the samples in 60-70 hours comprised 4 mg/cm^2, then for a prolonged period of time their weight was practically unchanged. In some of the samples there was a reduction in weight and in 200 hours the losses in weight comprised 4.5-6 mg/cm^2. Here some of the samples after 100-200 hours of testing lost their strength and were destroyed following a weak mechanical effect, others preserved their initial form and remained sufficiently strong after 300 hours of testing.

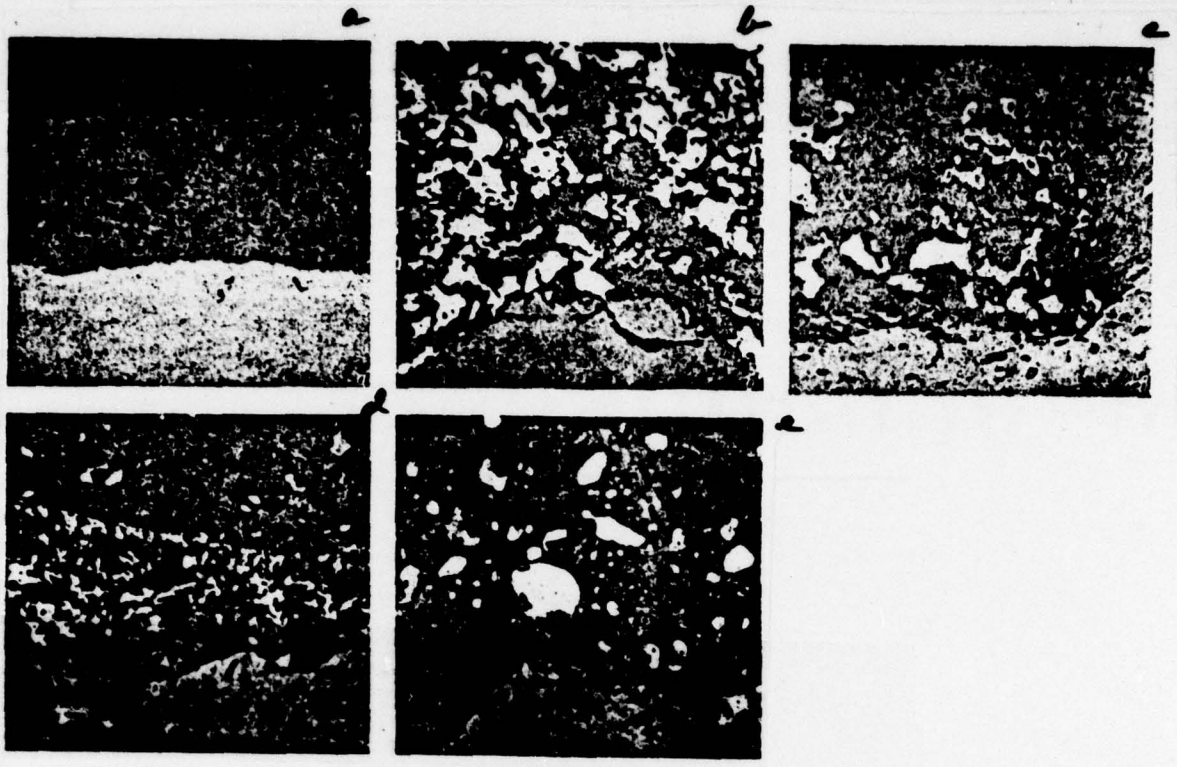

Figure 2. Microstructure of silicon carbide on a nitride binder without a coating and with a coating before and after testing at 1500° in the air.
a - without a coating, 240 hours, 1500°; b, c - with coating No 6: b - before testing, c - after 360 hours; d, e - with a double-layer coating: d - before testing, e - after 500 hours.

On their surfaces there was a glass-like film with a thickness of 10-20 μm. The glass-like film was permeated with fine (up to 2 μm) pores, sometimes large pores and even cracks were encountered in it (Figure 2,a).

Consequently, silicon carbide on a nitride binder is not sufficiently stable in air, and the scattering of data on heat tolerance and strength which was observed in it can apparently be explained by the inhomogeneity of the material.

SELECTION OF THE GLASS. From the point of view of obtaining glasses which are highly viscous and noncrystallizing at high temperatures there is interest in the areas of homogeneous glass in two ternary systems: SiO_2—Al_2O_3—TiO_2 and SiO_2—Al_2O_3—ZrO_2 [5].

We synthesized small amounts of glass of the composition given in Table 1.

Table 1

Compositions of Glass

(1) №№ стекол	Содержание компонентов, вес. % (2)			
	SiO_2	Al_2O_3	TiO_2	ZrO_2
1	70	5	—	25
2	80	5	—	15
3	85	5	10	—
4	90	5	—	5
5	90	5	5	—
6	95	2.5	2.5	—
7	95	2.5	—	2.5

Key: (1) No of glass; (2) Content of components, wt.%.

The glass was synthesized in the following manner. The finely dispersed components of the charge were mixed together with the addition of a small amout of water and dextrin. From such a freshly prepared paste rods 30 mm long and 3 mm in diameter were formed by the method of extrusion from glass tubes. The rods were dried, fired in the air at 1400° for 3 hours, and then subjected to melting in an electric arc with graphite electrodes. The glass was extracted in the form of globules of irregular form with a diameter of 6-8 mm. The quality of the glass was evaluated based on its crystallizing capacity in air at 1500° for 3 hours. Under these conditions all the glass, with the exception of composition No 6, was crystallized. The absence of crystallization in glass of composition No 6 was confirmed by electron-microscopic investigation. Glass No 6 was taken as the glass binder for the synthesis of the coatings. The amount of glass necessary for the investigation was

fused in a vacuum-compression high-frequency induction furnace in a medium of argon (10 atm(gage)) at 1800°.

PRODUCING THE COATINGS. Glass-silicide coatings were applied to samples made from SiC on a nitride binder with dimensions of 8 x 8 x 4 mm; the method of obtaining the coatings and the methods of studying their properties are described in [4]. The coatings were partially fused in a furnace with a tungsten heater in the temperature range of 1600-1800° in a medium of argon for 1-10 min. As a rule the coatings were applied in 2-3 stages. The overall thickness of the layer was 150-300 μm.

THE COATINGS AND THEIR PROPERTIES. The compositions of the tested coatings are given in Table 2.

Table 2

Composition of the tested coatings and their KTR

(1) №№ покрытий	(2) Содержание компонентов, вес. %		(4) КТР · 10^7, 1/град.
	стекло № 6 (3)	MoSi$_2$	
1	100	—	7
2	90	10	20
3	70	30	45
4	50	50	63
5	30	70	85
6	10	90	86
7 (5) подслой	30	70	—
7 (5) покровный слой	70	30	—
8 (6) подслой	10	90	—
8 (6) покровный слой	70	30	—

Key: (1) No of coating; (2) Content of components, wt.%; (3) Glass No 6; (4) KTR · 10^7, 1/deg; (5) sublayer; covering layer; (6) sublayer; covering layer.

It was found that the glass was fused on the SiC with a nitride binder at 1600° in 1-10 min with the formation of a film which was thick and firmly adhered to it. At temperatures higher than 1600° the glass was agglomerated into drops. Precisely the same thing happened to glass with an additive of bentonite (2 parts by weight

over 100%). The coatings of compositions Nos 2 and 3 with 10 and 30% $MoSi_2$ under these conditions were formed with the formation of vitrified nonporous layers, firmly adhered to the main layer. With an increase of the content of $MoSi_2$ the obtaining of a thick coating became difficult. In order to obtain a nonporous coating with 50% $MoSi_2$ it was necessary to apply 4-5 layers for the reliable covering of the pores of the preceding layers. Nonporous coatings of compositions Nos 5 and 6 could not be obtained. Double-layer coatings Nos 7 and 8 were obtained which were vitrified and nonporous.

Coatings Nos 2-8 were tested for heat tolerance at 1500° in a quiescent atmosphere of air. The results of the tests are given in Figure 1, 3-7. When defects appeared in the tests they were terminated. A characteristic for all the coatings was a certain increase in the weight of the samples in the initial period of testing and the vitrification of their surface layer. In this case the porosity of the coatings was reduced. In 50 hours of testing at 1500° the gain in weight comprised 0.5-4.0 mg/cm^2. The gain in weight of the samples with a coating increased along with an increase in the content of $MoSi_2$ in the coating.

Based on heat tolerance at 1500° the best results were given by the double-layer coating of composition No 8. In 500 hours of testing at 1500° no defects were revealed in it. The gain in weight comprised ~2.0 mg/cm^2.

The KTRs of glass and SiC on a nitride binder were determined on an Ulbricht dilatometer in the area of 20-800°, and for glass-silicide coatings they were calculated approximately using the formula of Kingeri [6] (Table 2). The KTRs of the coating of composition No 3 and SiC on a nitride binder were practically equal.

The microstructure of the coatings before and after the tests are shown in Figure 2. It is evident that in the process of testing the surface layer of the coatings was vitrified. After 360 hours its thickness on coating No 6 was 20 μm, and after 500 hours on the double-layer coating No 8 - 50 μm. In the process of prolonged testing the amount of the glass phase in the coatings is increased. X-ray analysis showed that there were no phase changes

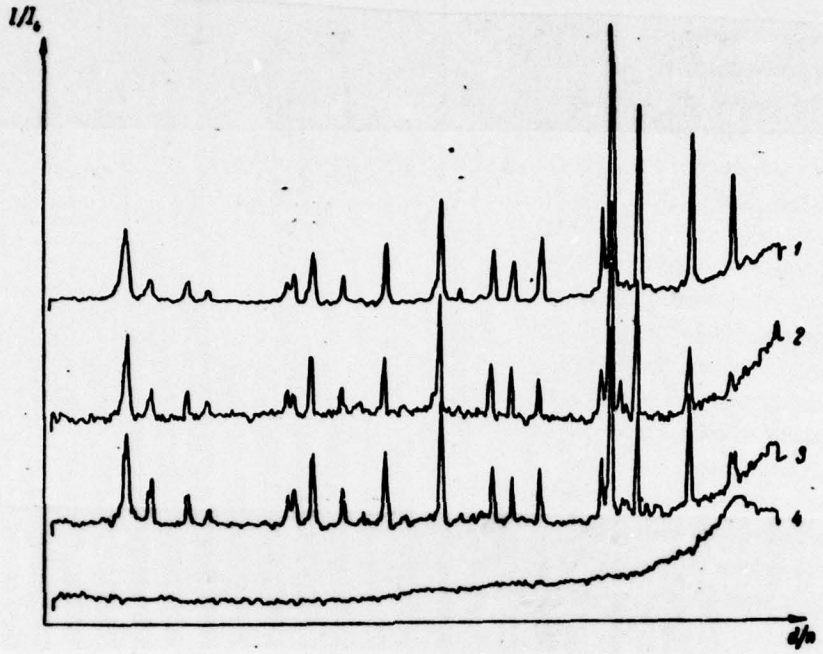

Figure 3. Difractograms.
1 - $MoSi_2$; 2 - samples of composition No 6 before testing;
3 - sample of composition No 6, 1500°, 100 hours; 4 - glass.

in coating No 6 (Figure 3).

For the purpose of clearing up the processes which take place in glass silicide coatings at high temperatures investigations were made of samples of the material of coating No 6 with the help of micro-X-ray spectral and electron-microscopic methods. The samples for the investigation were obtained in a medium of argon at 1600° for 2 min and were held at 1500° in the air for 100 and 360 hours.

Preliminary data were obtained on a "Cameca MS-46" device on the diffusion in glass-silicide coatings at 1500° of molybdenum from $MoSi_2$ into the glass binder and oxygen into the particles of $MoSi_2$.*

* Micro-X-ray spectral analysis performed by Yu. P. Udalov.

Figure 4. Distribution of Mo and O in the zone of contact of $MoSi_2$ with glass after 360 hours of oxidation in air at 1500°.
a - inside the sample; b - in the surface layer of the sample.
Key: (1) Distance from the phase interface, μm; (2) Concentration; (3) Film.

Figure 5. Electron-microscopic image of a sample of composition No 6 after testing in air at 1500° for 100 hours.

Qualitative analysis showed that in the process of formation of a coating diffusion processes on the boundary SiC—glass, $MoSi_2$—glass are practically absent. In the samples held for a prolonged time at 1500° in air the penetration of molybdenum from particles of $MoSi_2$ into the glass and oxygen into the particles

of $MoSi_2$ was detected. Figure 4 shows the distribution of molybdenum and oxygen in the zone of contact of particles of $MoSi_2$ with glass on the surface of the investigated sample and inside of it (at a distance of 1 mm from the surface). It is evident that inside of the sample (Figure 4, a) the molybdenum diffuses from $MoSi_2$ into the glass to a depth of 6 μm. Oxygen diffuses into the particles of $MoSi_2$ to a depth of 2 μm. In the surface layer of the sample (Figure 4, b) the oxygen diffuses into the particles of $MoSi_2$ also to a distance of 2 μm, however, in the glass which is enveloping the particles of $MoSi_2$ in the surface layer molybdenum was not detected. This can be explained by the volatilization of molybdenum oxide from the surface layer. Inside of the sample molybdenum oxide is evidently dissolved in the glass.

Electron-micorscopic analysis showed that after 100 hours of testing in air at 1500° the glass phase is not crystallized. In Figure 5 grains of $MoSi_2$ surrounded by homogeneous glass are evident. The surface of the grains is rough, which can be explained by the diffusion in the surface layer glass of the particles of $MoSi_2$ and by the presence in them of micropores or microcracks.

Thus glass silicide coatings were obtained which were intended for protecting silicon carbide on a nitride binder from oxidation in a medium of air. Coatings with a thickness of 0.1-0.2 mm protect SiC at 1500° for 500 hours without the appearance of defects. The heat tolerance of samples with coatings comprises 0.5-4.0 mg/cm^2 for 500 hours. It was demonstrated that at high temperatures there is an interaction of components of the coating with one another and with the oxygen of the air.

BIBLIOGRAPHY

1. С. А. Добролеж, С. М. Зубкова, В. А. Кравец, В. З. Смушкевич, К. Б. Толпыго, И. Н. Францевич. Карбид кремния. Киев, 1963.
2. И. С. Кайнарский, Э. В. Дегтярева. Карборундовые огнеупоры. Харьков, 1963.
3. C. G. Rose, J. C. Andersen, пат. США № 3252827, 1966.
4. М. В. Савонова, Г. Н. Комарова, в сб.: Жаростойкие покрытия, изд. «Наука», М.—Л., стр. 125, 1965.
5. A. Bodgar, L. Doney, Glas. Ind., 21, № 7, 309, 1940.
6. У. Д. Киппери. Введение в керамику. Стройиздат, М., 1964.

DISTRIBUTION LIST

DISTRIBUTION DIRECT TO RECIPIENT

ORGANIZATION		MICROFICHE
A205	DMATC	1
A210	DMAAC	2
B344	DIA/RDS-3C	8
C043	USAMIIA	1
C509	BALLISTIC RES LABS	1
C510	AIR MOBILITY R&D LAB/FIO	1
C513	PICATINNY ARSENAL	1
C535	AVIATION SYS COMD	1
C591	FSTC	5
C619	MIA REDSTONE	1
D008	NISC	1
H300	USAICE (USAREUR)	1
P005	ERDA	1
P055	CIA/CRS/ADD/SD	1
NAVORDSTA (50L)		1
NASA/KSI		1
AFIT/LD		1

ORGANIZATION		MICROFICHE
E053	AF/INAKA	1
E017	AF/RDXTR-W	1
E404	AEDC	1
E408	AFWL	1
E410	ADTC	1
E413	ESD	2
	FTD	
	CCN	1
	TQIS	3
	NIA/PHS	1
	NICD	2